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Experimental and numerical analysis of microstructure damage in silica filled epoxy

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Abstract

The epoxy resin-based systems with silica filler are widely used in many products like medium and high voltage electrical components due to its very good dielectric and mechanical properties. Such products require to operate in harsh environments which may activate the process of formation and propagation of the cracks within the resin material. The cracking phenomenon contributes also to manufacturing problems. The epoxy based parts are very often produced by casting during which (post)curing cracking may appear.

In order to better understand the cracking phenomena of epoxy resin there is a need to investigate microstructural damage. The work presented in this paper includes both experimental and numerical analysis of microstructure crack initiation and propagation in silica filled epoxy. Basic information about epoxy resin-based systems and its applications are presented. Next, basics of the fracture mechanics with description of available numerical approaches are described. The numerical simulations were prepared for Representative Volume Element (RVE) which was obtained using home-made tool for image digitalization. Experimental analysis consist of in-situ tensile tests and microstructural observations with Scanning Electron Microscope (SEM). At the end a summary with conclusions related with prepared numerical analysis and experiments is included. The presented research of the damage of silica/epoxy composite confirms that analysis of the epoxy resin microstructural damage is not trivial and further study is required for its better understanding.

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1. Introduction

The epoxy resin-based systems are widely used in industry and technology. Depending on the desired properties of the composite, various fillers can be added to the polymer matrix. By this the required properties such as mechanical, chemical, thermal can be obtained. One of the major issue of epoxy resin-based systems is cracking. Due to the brittleness of some commonly used materials, e.g. epoxy resin with silica, following questions are still open:

- What is the filler influence on the fracture strength of the composite?
- Can we anticipate the moment of crack initiation and its behavior?
- Do analytical models approximate well the process of the cracking of powder composites?

Nowadays, development of computers and simulation software allow modeling of various physical phenomena, such as flow, statics or cracking with an increasing precision. Professional computational packages have very powerful modules for the calculation of the destruction of materials, using a various techniques. So, it seems natural to use it for better understanding of the fracture mechanics of the epoxy resin-based systems. However, as always, a numerical approach should be confirmed by the result of experiments.

2. Epoxy filled composite

2.1. Application

In last 30-40 years epoxy resins have found widespread applications in the manufacturing of medium and high voltage electrical components. Such products include switchgears, breakers, instrument and distribution transformers, as well as bushings and insulators. Many of these applications are required to operate in harsh environments, for example outdoor applications in regions exposed to intensive sunlight high humidity or excessive thermal conditions. Typically, the design of products cast in epoxy material contains metal inserts with vastly different properties, especially thermal expansion coefficient and mechanical stiffness. Such demanding requirements for operating conditions as well as thermo-mechanical interaction with embedded components may activate the process of formation and propagation of cracks within the resin material. The cracking phenomenon affects not only the electrical apparatus in service conditions, but contributes also to manufacturing problems since, in some cases, a quite noticeable part of production must be scraped due to (post)curing cracking (Nowak et at., 2009).

2.2. Material properties

By adding a filler, like silica, to the polymer matrix, physical, mechanical and thermal properties of the composite are changed. In simplified way, the properties of composite can be determined by the rule of mixtures based on volume fractions of components in the composite. However, in that case the filler size, its shape and the strength of the filler/matrix interface may have influence on the mechanical properties. In order to analyze the microstructural cracking of epoxy-based composite the basic mechanical properties have to be determined: Young's modulus and critical strain energy release rate. In case of Young's Modulus, traditional tension test can be used. In case of Critical Strain Energy Release rate, the number of approaches is limited. In this case special samples were prepared and tested using Optical Crack Tracing (OCT) technique by Fraunhofer Institute in Germany. Values of those properties are presented in next sections of this paper.

3. Fracture mechanics

3.1. Basics

Fracture mechanics is still a new, developing approach of materials strength (Anderson, 2005). In the opposite to classic materials strength approaches, where it is assumed that the material is ideal and does not have any imperfections, in fracture mechanics it is assumed that there are some discontinuities in the material. As a consequence, the material strength depends on three parameters (applied load, crack size, fracture resistance) instead of two (applied force, material resistance) used in classical mechanics (Wei, 2005). Three cracking modes can be distinguished: opening, in-plane shear, out-of-plane shear. It should be noted that usually there is a need to deal with mixed types of cracking modes (Ochelski, 2004). Each of the above-mentioned methods corresponds with stress field, which is:

$$\sigma_{uj}^{T} = \frac{K_T}{\sqrt{2\pi r}} f_{ij}^{T}(\theta) \tag{1}$$

where:
σ - stress,
i, j - x, y or z coordinates
K – stress intensity factor,
T – cracking mode (1, 2 or 3),
r, θ – polar coordinates placed in the crack tip
f – functions dependent on *θ*

In an isotropic, elastic material, functions *f* take the form of following equations:

$$\sigma_{xx} = \frac{K_T}{\sqrt{2\pi r}} \cos\left(\frac{\theta}{2}\right)\left[1 - \sin\left(\frac{\theta}{2}\right)\sin\left(\frac{3\theta}{2}\right)\right] \tag{2}$$

$$\sigma_{yy} = \frac{K_T}{\sqrt{2\pi r}} \cos\left(\frac{\theta}{2}\right)\left[1 + \sin\left(\frac{\theta}{2}\right)\sin\left(\frac{3\theta}{2}\right)\right] \tag{3}$$

$$\tau_{xy} = \frac{K_T}{\sqrt{2\pi r}} \cos\left(\frac{\theta}{2}\right)\sin\left(\frac{\theta}{2}\right)\cos\left(\frac{3\theta}{2}\right), \tag{4}$$

and the stress distribution around the crack peak can be described as shown in Fig. 1.

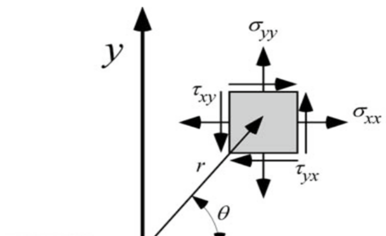

Fig. 1. Stress distribution around the crack peak.

One can notice that if $r \rightarrow 0$, then $\sigma \rightarrow \infty$. This means that each crack has a singularity at the peak.

There are a lot of approaches for fracture mechanics. The classic ones are based on time-independent crack behaviour (Linear-Elastic or Elastic-Plastic Fracture Mechanics) and on material isotropy. But the new methods, like time-dependent or based on material microstructure, are still being developed on numerical simulations market.

3.2. Approaches for analysis of microstructure damage

In the literature a number of numerical approaches that can be used for analysis of cracking in composites can be found. The most popular methods are Contour Integral (CI), Element-based cohesive behaviour (EBCB), Surface-based cohesive behaviour (SBCB), Virtual crack-closure technique (VCCT) and Extended Finite Element Method (XFEM). Each of these methods has advantages and disadvantages, but two of them may be particularly helpful during the analysis of microstructural damage in epoxy resin-based materials. One of them is Virtual-Crack Closure Technique (VCCT) which is one of the newest approaches, based directly on brittle fracture mechanics equations (Krueger, 2014). It is a very powerful technique for modelling brittle fracture and delamination. However in these days the most promising technique for modelling damage in the composite materials is eXtended-Finite Element Method, developed by Belytschko and collaborates (Belytschko et at., 1999). XFEM method is based on the partition of unity instead of crack propagating along the nodes. Application of this technique and obtained results are presented in the paper.

4. Numerical analyses of 2D sample

4.1. Image digitalization of RVE

For numerical analyses a 2D Representative Volume Element (RVE) was generated on the basis of a real microstructure analysed by scanning electron microscope (SEM). In such a SEM-image of the real microstructure, the grey scale values of the different pixels were evaluated by the in-house developed software tool. . By defining a threshold value, the polymer matrix and silica filler area could be differentiated automatically. Afterwards, the image was converted to a 2D Finite Element model. With this method, the influence of phase distribution and fraction on the mechanical properties and the failure behaviour can be taken into account. In the first analyses, in order to reduce the required computation time, the numerical model has been simplified as can be seen in Fig. 2. RVE has dimensions of 100x100 μm.

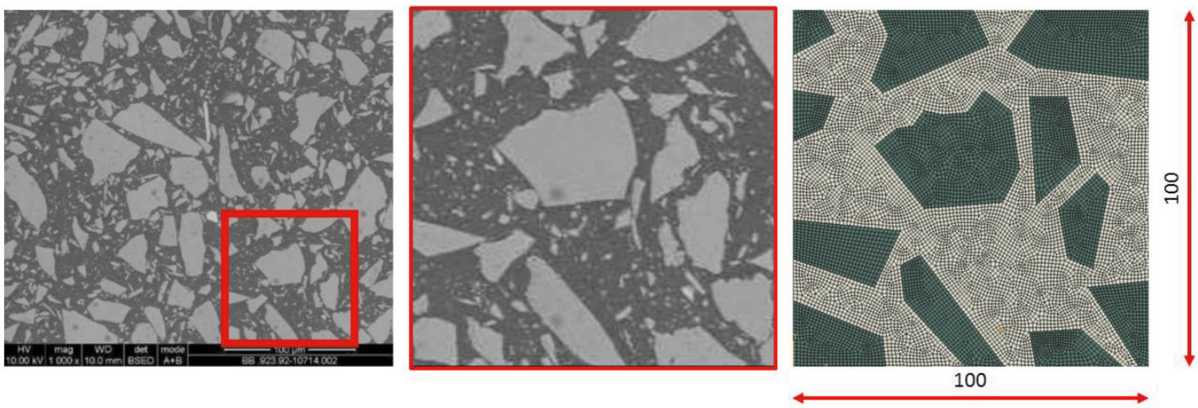

Fig. 2. Exemplary SEM image of the micro-silica filled epoxy composite (left and middle) and 2D FE model (right).

4.2. Material properties and boundary conditions

According to experiments results, the following mechanical properties of the epoxy resin and silica have been used in simulations (Table 1).

Table 1. Material properties of matrix and particle.

Material	Young's Modulus [MPa]	Poisson's Ratio [-]
Matrix (epoxy resin)	1200	0.37
Particle (fused silica)	70000	0.17

Maximal Principal Stress (MaxPS) was chosen for Fracture Initiation Criterion and the value that cause the failure was set to 60 [MPa]. Adhesion was taken into account during the simulation. Properties that have been assumed during the simulation are presented in Table below:

Table 2. Critical strain release rate and calculated K_{eff} and T_{ult}.

G_C [mJ/mm2]	K_{eff} [mJ/mm2]	T_{ult} [MPa]
0.001	1600000	40

The values K_{eff} and T_{ult} have been calculated using the following equations:

$$K_{eff} = \frac{2G_C}{\delta_{ratio} \cdot \delta_{fail}^2} \tag{5}$$

$$T_{ult} = \frac{2G_C}{\delta_{fail}} \tag{6}$$

where:
G_C – critical strain release rate
T_{ult} – nominal stress at damage initiation
K_{eff} – effective stiffness
δ_{ratio} – constant. If the value is unknown $\delta_{ratio} = 0.5$
δ_{fail} – the value dependent of the mesh density. $\delta_{fail} = 0.05 \cdot l_{cz}$, where l_{cz} – average element size in cohesion connection.
The following boundary conditions have been assumed and are presented in Fig. 3.

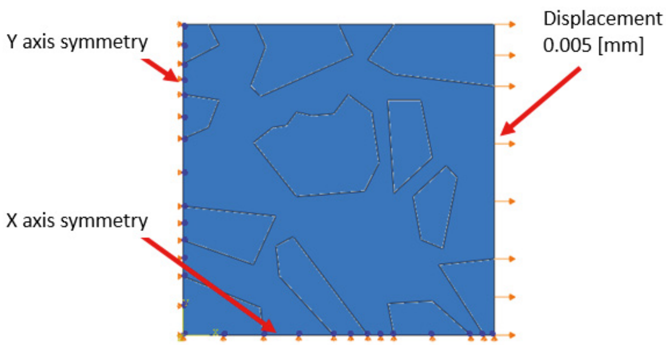

Fig. 3. Boundary conditions.

4.3. Results

As the results the stress-strain curves, stress distribution and crack initiation locations have been obtained. Selected results are shown below. Stress-stress curve is presented in Fig. 4.

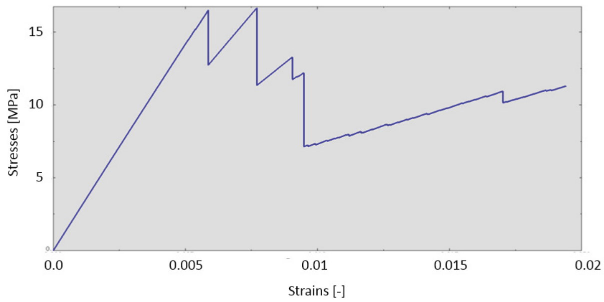

Fig. 4. Stress-strain curve.

Peaks visible in Fig. 4. above indicate delamination between the matrix and silica filler, during which a portion of energy was released. The stress distribution for presented case and the crack initiation locations marked with red circles are presented in Fig. 5.

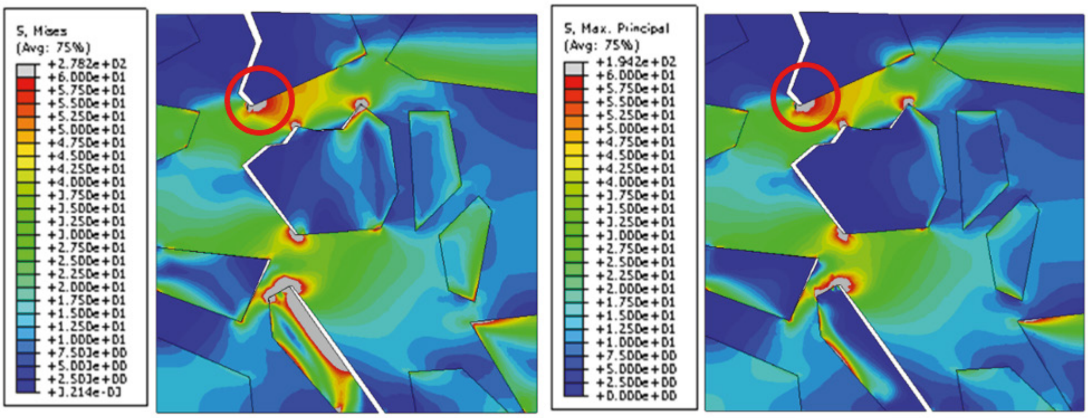

Fig. 5. Stress [MPa] distribution and selected crack initiations (red circles) at the end of analysis.

These simulations results would indicate that initiation of epoxy resin material damage probably takes place in the interface as a result of the loss of interface strength and delamination.

5. Experimental investigation

5.1. Experimental methods review

There are many methods of testing material resistance to cracking. However, they are related to metals. The problem of cracking of polymers and composites is relatively new. Accordingly, the number of standards for the determination of fracture toughness for these materials is limited, although in recent years, many research centers have addressed this issue. The main problem for testing the properties of the polymers is their creep, which in many cases cannot be overlooked in relation to the standards would be accurate. Typical experimental methods (samples) that can be used for polymers are:

- Compact Tension (CT)
- End Notched Flexure (ENF)
- Edge Crack Torsion (ECT)

Dobrzanski (2002) reports that the composite powder can be regarded as macroscopically isotropic materials. However, at the micro level, , the following phenomena could appear:

- discontinuous stress distribution caused by different Young's modules and Poisson coefficients of matrix and filler materials,
- initiation of cracks in different places depending on the shape of inclusions and the interface parameters,
- turning the crack tip in the direction of, or to a particle,
- delamination of the particles.

The above-mentioned factors determine the fracture toughness of the composite, and in these days determination of all composite properties (like adhesion on the interface) is very challenging.

5.2. Scope of the experiment

The aim of the experiment was to determine the tensile strength of the composite and to observe the microstructure in order to analyze the process of crack initiation and propagation. The experiment was distinguished into two stages. In the first one, using a Scanning Electron Microscope, the microstructure of the composite was obtained. In the second one, the tensile strength of the samples has been determined using Microtest 5000 tensile machine.

5.3. SEM observation and tensile strength tests

The tests were performed using Microtest 5000 tensile machine from Gatan Company. One test was carried out in the chamber of a scanning electron microscope with the observation of the specimen during testing, what has been shown in Figure 6.

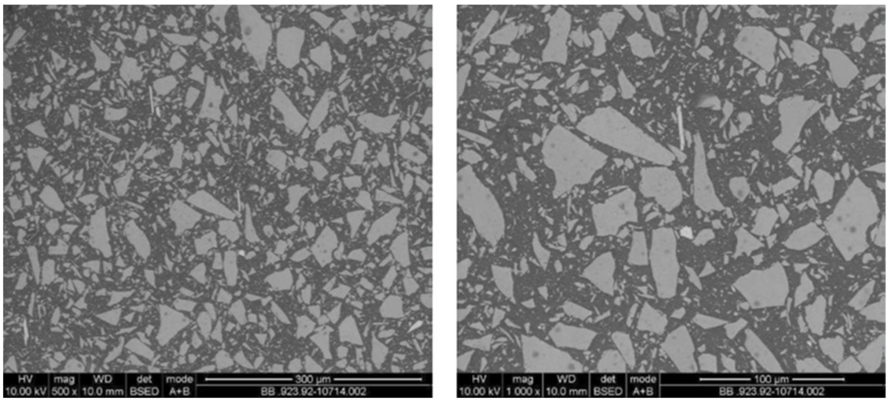

Fig. 6. The microstructure obtained using the BSE detector (Backscattered Electron).

Pictures of the broken sample after the tensile tests are presented in Fig. 7.

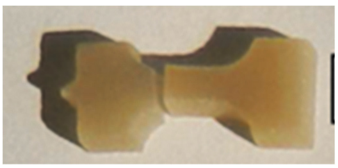

Fig. 7. Sample after tensile test.

All samples broke at the same place, and the cracks have similar shape. Performed observations using SEM revealed a complex microstructure of the composite silica-filled (Fig. 8.). Observing the process of initiation and propagation of cracks using a scanning electron microscope is very demanding as it is a very rapid process. The resulting images show a breakthrough that in the tested composite crack propagates in the warp and often runs at the interface between the matrix and the inclusions.

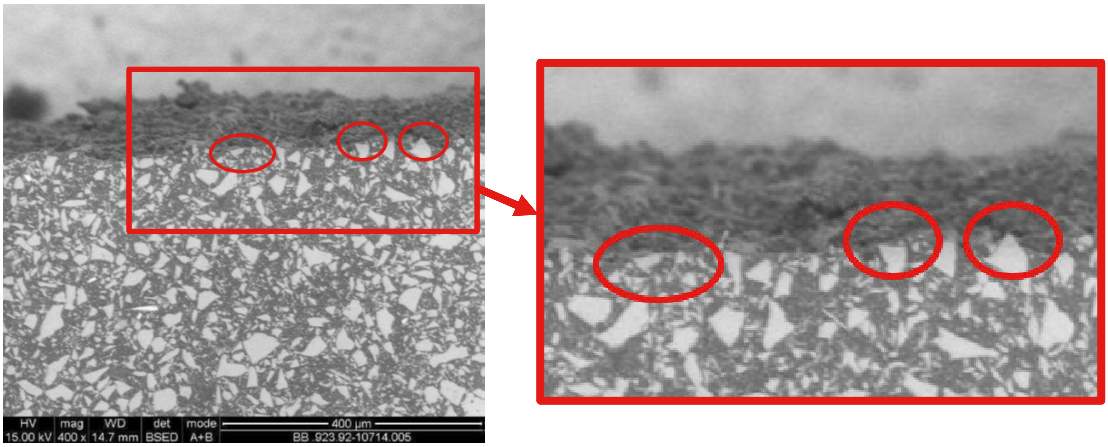

Fig. 8. Fracture surface and microstructure of the composite with marked areas of delamination.

6. Summary

Silica filled epoxies are commonly used in power products mainly as insulating materials to encapsulate components on high electrical potential , owing to their very good dielectric properties. Power products require to operate in harsh environments, for example outdoor applications in regions exposed to intensive sunlight, low temperatures, or excessive thermal extremes. These requirements may activate the process of formation and propagation of the cracks within the resin material. The cracking phenomenon affects not only the electrical apparatus in service conditions, but contributes also to manufacturing problems. In order to understand better the cracking phenomena of epoxy resin there is a need to investigate microstructural damage. The work presented in this paper includes both experimental and numerical analysis of microstructure crack initiation and propagation in silica filled epoxy. For numerical analysis of damage of the epoxy resin Extended Finite Element Method (XFEM) has been used. The numerical simulations were prepared for Representative Volume Element (RVE) which was obtained using home-made tool for image digitalization. Further study was conducted with experiments – in-situ tensile tests and microstructural observations with Scanning Electron Microscope (SEM). Microstructural analysis and numerical simulations indicate the fact that initiation of epoxy resin material damage probably takes place at the interface as a result of cracking and loss of silica/matrix bond. This results in weakening of the epoxy resin microstructure in this area which leads to further structural degradation, consisting in the propagation of matrix cracks and, as a result, complete damage of the composite structure. The presented research of silica/epoxy composite damage confirmed that analysis of the epoxy resin microstructural damage is not a trivial and further study is required for its better understanding.

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